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Abstract

PYROMETALLURGICAL AND PYROCHEMICAL FUEL PROCESSING

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High-temperature fuel reprocessing methods are being developed in order to achieve rapid recycle of discharged reactor fuel in as direct a manner as possible. With these processes, low fuel recycle costs are expected to result from minimum fuel inventory charges, avoidance or simplification of chemical conversions, small process volumes and, hence, compact processing equipment, direct production of solid wastes, absence of severe critical mass restrictions, and the possible retention of valuable fuel alloying additives. Because of these characteristics, pyrometallurgical processes can be employed for recovery of fast reactor fuels in processing plants located on the reactor site.

A processing facility adjacent to the Second Experimental Breeder Reactor (EBR-II) will be used to demonstrate various pyrometallurgical processes in connection with recovery of EBR-II fuels. The first EBR-II fuel, an enriched uranium-fissium alloy, will be recovered by melt refining, a simple melting procedure by which fission products are removed by volatilization and oxidative drossing reactions. Plant equipment for melt refining 10-kilograms batches of fuel has been installed in the EBR-II Fuel Cycle Facility and is expected to be in operation before the end of 1964.

Two other processes are required in the EBR-II fuel cycle to (1) recover U-235 from melt refining crucible residues and (2) concentrate the plutonium bred into the uranium blanket material. In these processes, molten metals (e.g., zinc and magnesium) and molten halide salts are used as solvents. Both processes have been advanced into the pilot plant stage of development. Other

processes utilizing liquid metal and molten salt media are being developed for the recovery of fast breeder fuels containing plutonium. Metal, oxide, and carbide fuels can be accommodated in the liquid metal-salt processes.

At Hanford, a closed-cycle process using a molten chloride solvent, such as a lithium chloride-potassium chloride system, is being developed in connection with the Plutonium Recycle Program (PRP). The fuel, a mixture of uranium dioxide and plutonium dioxide, is dissolved by sparaging the molten chloride bath with hydrogen chloride or chlorine. Recovery of the uranium and plutonium from the melt, in forms suited to recycle to the reactor, can be achieved by: (1) electrodeposition of essentially plutonium-free uranium dioxide, (2) electrodeposition of solid solutions of uranium and plutonium dioxides, and (3) precipitation of essentially uranium-free plutonium dioxide.

Reactions of gaseous reagents with uranium oxides and carbides are being used in fuel reprocessing studies at Atomics International. The objectives of the processes are limited to repair of radiation damage and re-enrichment of the fuel. Uranium oxide fuels are pulverized for refabrication by alternate oxidation with oxygen or air and reduction with hydrogen. One process for uranium monocarbide fuels consists of gaseous oxidation followed by carbothermic reduction of the oxide to regenerate the carbide. Another consists of gaseous nitridation to produce a uranium nitride-carbon mixture followed by reconversion to the monocarbide by heating the mixture in a vacuum to above 1500C. A third process, which, in contrast to the gaseous-reaction processes, provides significant removal of fission products, involves electrolytic dissolution of the carbide in a molten chloride bath. The metallic uranium product is dissolved in mercury and reconverted to the carbide by distillation of the mercury in a hydrocarbon atmosphere.